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Properties Of Ammonium Nitrate-Urea-Water Hygroscopic Reagent For Warm Fogs

by

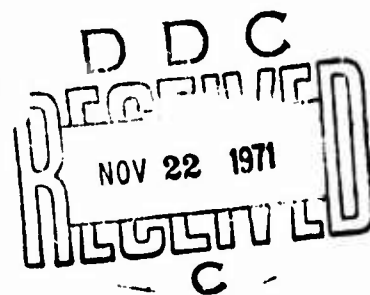
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W. J. Moran, RADM, USN Commander
H. G. Wilson Technical Director

FOREWORD

The search for suitable hygroscopic agents to use in fog and cloud modification has led many investigators down many diverse and sometimes strange pathways. The compound described herein is quite suitable for the purpose, in that it is cheap, safe, nontoxic, and indeed, beneficial and easy to handle. It is adequately hygroscopic so that it works rapidly and at present seems to us the best available agent.

This study was supported by AIRTASK A370-5401/216C/OW 3712-0000, Atmospheric Applications from AIR-540, Naval Air Systems Command.

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CONTENTS

Introduction 1

Hygroscopicity 1

Availability and Cost 2

Handling 2

Ecological Effects 2

Chemical Properties 3

Sensitivity 9

Drop Growth 10

Conclusion 19

Appendix: Drop Size Calculations
 (Equation, Values, Computer Program, and
 Sensitivity of Equation) 20

References 26

INTRODUCTION

Warm fog can be cleared by several different approaches. The fog can be evaporated by using large amounts of energy; the fog can be collected in filters; foggy air can be mixed with drier air to evaporate the droplets; the fog can be caused to coalesce by charging some of the droplets; or the fog droplets can be dried up and swept out by spraying hygroscopic particles onto the top of the fog.

The Earth and Planetary Sciences Division of the Naval Weapons Center began fog dispersal studies in 1963 using a saturated solution of ammonium nitrate in water to adsorb water vapor and dry out the fog. Two hundred gallons of this solution were sprayed from agriculture-type spray aircraft flying as slowly as possible in a tight turn. As the initial studies met with considerable success, the efforts were continued on a very modest scale over the next several years. In 1968 the studies were expanded and a series of potential fog dispersal agents were systematically screened. The evaluation criteria included hygroscopicity, availability, cost, ease of handling, and environmental effects. The agents screened included particulate aerosols, liquids, and powders. Both the ammonium nitrate solution mentioned above and solid, finely divided urea were tested with promising results.

During the latter part of the testing program, combinations of hygroscopic agents were tried. The most promising of these, ammonium nitrate, urea, and water in approximately equimolar ratios was selected as the agent of choice. Although the agent has been used during the past two years with gratifying results, the purpose of this paper is not to discuss these results. The test results are presented in Naval Weapons Center publications, Foggy Cloud I (Ref. 1), Foggy Cloud II, in press, and Foggy Cloud III, in preparation. The purpose of this paper is to summarize the properties of the agent.

HYGROSCOPICITY

The first criterion, high hygroscopicity, made the ammonium nitrate-urea-water solution a prime candidate for evaluation. The utility of this material, as demonstrated in field tests, has been substantiated by two independent laboratory investigators as reported below.

Berg¹ stated that "when seeding is done at the desired ceiling, the material may be expected to sorb water vapor, to, say, 100 times its own volume". He further

¹Report from T. G. Owe Berg, Inc. to the Naval Weapons Center, 1 October 1969. Purchase Order No. N66001-70-6020Y0372.

stated in the same report that "80% of the growth by volume in 30 minutes took place within the first 15 minutes of exposure to the moist air". These conclusions are to be expected from the vapor pressure of 7.9 mm of Hg at 25.8°C for the undiluted agent as reported in Ref. 1, Appendix C. The vapor pressure approaches asymptotically that of pure water as the drop grows. At a relative humidity of 99% the asymptote is at a volume almost 50 times that of the original droplet. Further growth will be by coalescence or, at supersaturation, the drops will grow as a pure water drop by diffusion.

Berg compared the ammonium nitrate-urea-water material with three other potential agents. These included a finely divided sample of Morton Corp. (Hutchins Plant, 200 fine) sodium chloride, a finely divided sample of Leslie Corp. sodium chloride, and ground calcium chloride (an unknown amount of water present) of Deep Water Chemical Co. He concluded with "this material alone could be seriously considered for fog dispersal by sorption of water vapor".

AVAILABILITY AND COST

The material readily meets the second and third criteria, as both ammonium nitrate and urea are widely used as fertilizers and are commercially available in solution in approximately the same concentration as this hygroscopic agent. It is therefore easily obtainable at a cost of approximately five cents per pound.

HANDLING

The fourth criterion, ease of handling, was readily met. As the material is liquid, it is readily transferred from tank trucks to bulk storage to aircraft or other delivery vehicle. It may be stored in bulk containers for periods in excess of a year without deterioration, the only requirement being that its temperature be held above 13.7°C. When placed in a delivery vehicle, its high heat capacity keeps it in a liquid state amply long for normal operations.

ECOLOGICAL EFFECTS

The fifth criterion, ecological effects, was met when it was established that in no instance would the amount of nitrogen distributed over the test area annually exceed the maximum amounts used for agricultural purposes. The flora in the treated area has been carefully observed for the three test years.

The treated area has been delineated by placing strips of urea-sensitized filter paper throughout the target area and observing the limits of spread of the material. This test paper is prepared by dissolving 1 gram of *p*-dimethylaminobenzaldehyde in 12 ml of concentrated hydrochloric acid (sp. gr., 1.19). After dissolution, 900 ml of distilled water is added. The test paper, after being wetted in the solution and allowed to dry, is ready for use. It yields a bright yellow color upon contact with solution droplets.

In no instance during the test period have any deleterious effects upon the flora at the airport been noted. The effect of the agent on other materials, for example, wood, concrete, asphalt, and painted and metallic surfaces has been carefully noted. Although, as reported below, it does corrode mild steel, magnesium, brass, and cadmium, rapid washdown, that is, within two hours after exposure, has prevented any problems. No corrosion or deleterious effects have been noted with other materials.

CHEMICAL PROPERTIES

Liquid mixtures of ammonium nitrate, urea, and water containing these components in concentrations in the order of one mole of each are unusual "solutions" to say the least. This may be brought out by noting that, at 25°C, the molal (moles solute/1000 grams solvent) concentration in saturated aqueous solutions of very soluble materials such as ammonium nitrate, lithium chlorate, and sodium hydroxide is 39, 36, and 29 moles, respectively. At this temperature, a saturated aqueous "solution" of ammonium nitrate, urea, and water in equimolar proportions has a molal concentration of 177 moles. It appears that these liquid mixtures are best considered as complexes involving all three components. Such a view is supported not only by the high concentrations of ammonium nitrate and urea reached, but also by the fact that attempts to precipitate an ammonium nitrate-urea complex from these liquids have failed. Despite the fact that equimolar mixtures contain 140 grams (80 grams of ammonium nitrate and 60 grams of urea) of solids and only 18 grams of water, the water can be considered the solvent and the liquid mixture a solution on the grounds that water may be added to these mixtures indefinitely without a loss of homogeneity. For practical purposes, it is best to refer to these liquid mixtures as solutions. Reference is made to these solutions in terms of the solids to water ratio with the ratio of ammonium nitrate to urea understood to be equimolar. Thus a "9:1 solution" contains 9 grams of ammonium nitrate and urea and one gram of water.

In a continuing series of investigations (Ref. 1, Appendix C), data are presented on the physical properties of the ammonium nitrate-urea-water hygroscopic agent. These data that include solubility determinations, pH, solution densities and viscosities, and vapor pressure, *p*, measurements are given in Table 1 and Fig. 1 through 4.

With equimolar ratios of ammonium nitrate and urea, precipitation induced by cooling occurred in two stages. X-ray diffraction of the material precipitated in the first stage showed this to be ammonium nitrate. An X-ray diffraction pattern of the material from the second-stage precipitation showed this to be ammonium nitrate and

TABLE 1. Data on the Ammonium Nitrate-Urea-Water System.

Parts solids/ part water, by weight ^d	Composition, moles			Liquidus temperature, °C		pH	Vapor pressure, <i>p</i>		Density (vac) at 26°C, gm/cm ³	Viscosity at 26°C, centipoises
	Ammonium nitrate	Urea	Water	Primary	Secondary		mm Hg	°C		
12.442	1.60	1.60	1.00	25.20	22.80	4.80	1.381	...
11.664	1.50	1.50	1.00	5.9	26.0
10.116	1.30	1.30	1.00	20.25	16.60	5.20	6.9	25.8	1.372	14.6
8.942	1.15	1.15	1.00	16.30	13.00	12.6
7.776	1.00	1.00	1.00	13.70	9.50	5.50	7.9	25.8	1.361	10.5
6.998	0.90	0.90	1.00	10.50	7.60	5.29
6.221	0.80	0.80	1.00	7.40	...	5.19	9.1	26.0	1.346	...
4.666	0.60	0.60	1.00	1.50	...	5.50	9.5	25.7	1.328	5.8
3.110	0.40	0.40	1.00	11.3	26.1
1.555	0.20	0.20	1.00	14.7	26.0

^d Solids = 1 part urea + 1.33 parts of ammonium nitrate by weight.

urea. This two-stage precipitation results from the fact that maximum solubility in this system occurs with a composition containing a slight excess of urea. However, as temperatures of the solution used for project work are held above 25°C, no adjustment in ratios has been necessary (Fig. 1).

Studies of corrosion by the ammonium nitrate-urea-water system have been conducted. The method employed in these studies consists of placing a clean, weighed 1- X 3- X 1/16-inch specimen of the test metal in a 50-ml beaker containing 25 ml of the test solution. The beakers were covered to prevent changes in solution concentration. The samples were exposed for various time periods ranging from 95 to 600 hours.

Materials tested to date include magnesium alloy AZ31B, brass, mild steel, chromium plated steel, cadmium plated steel, cadmium, aluminum, and lacquers and enamels used for automobile finishes. Corrosion inhibitors, including thiourea, tribasic potassium phosphate, and ammonium iodide were tested. Ammonium iodide was found to be ineffective. Table 2 summarizes some results of the tests. Cadmium, although not shown in Table 2, is attacked vigorously, and cadmium plate apparently enhances the attack on steel. Although these studies are continuing, as mentioned above, during Foggy Cloud field tests and three additional major field experiments, no serious damage to aircraft or other metallic surfaces has been observed.

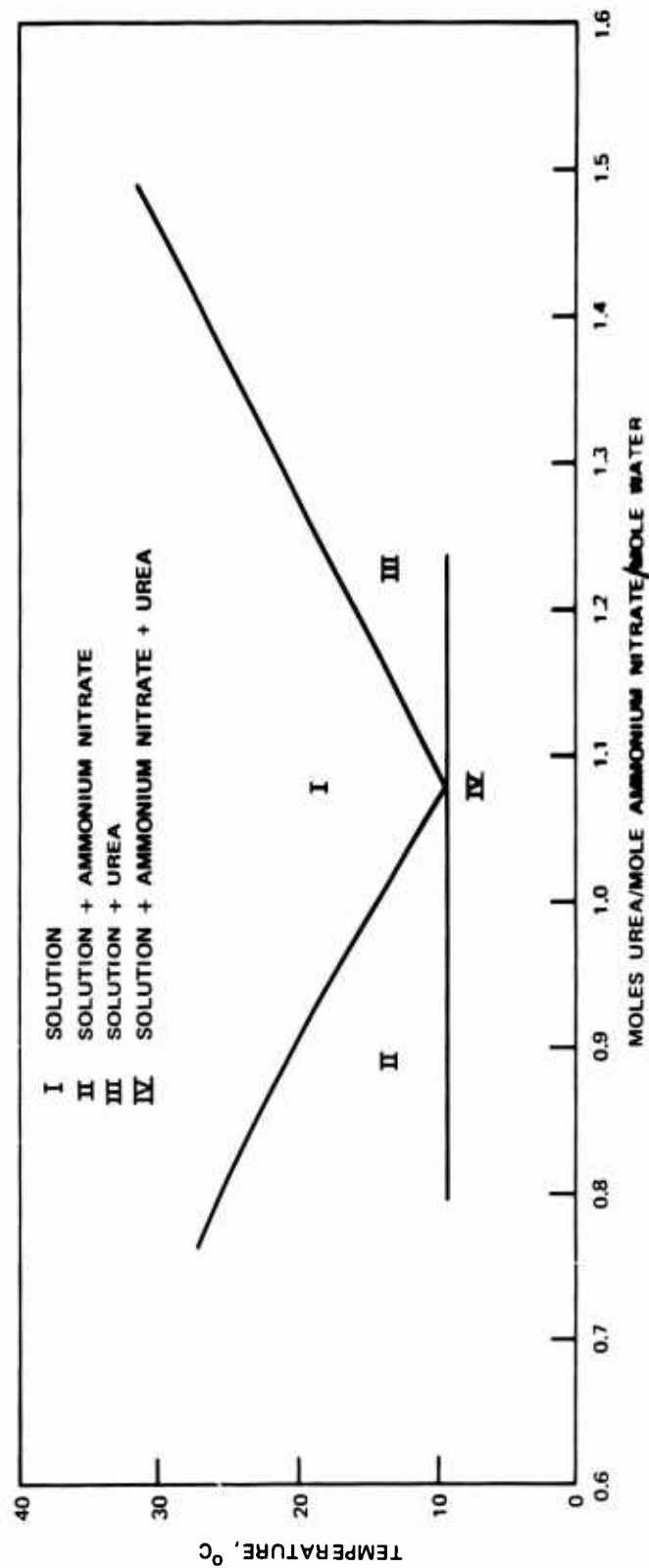


FIG. 1. Phase Diagram.

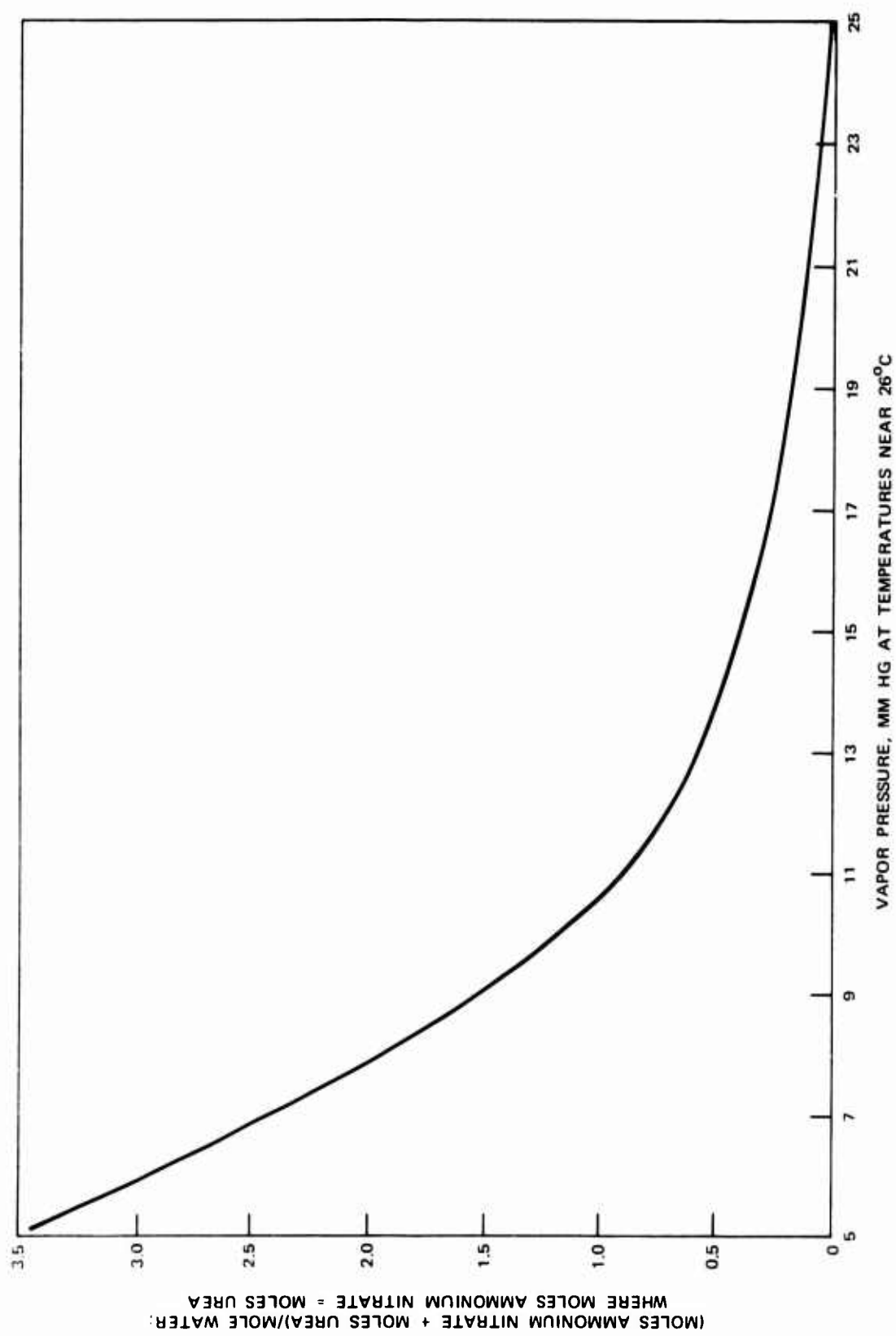


FIG. 2. Vapor Pressure.

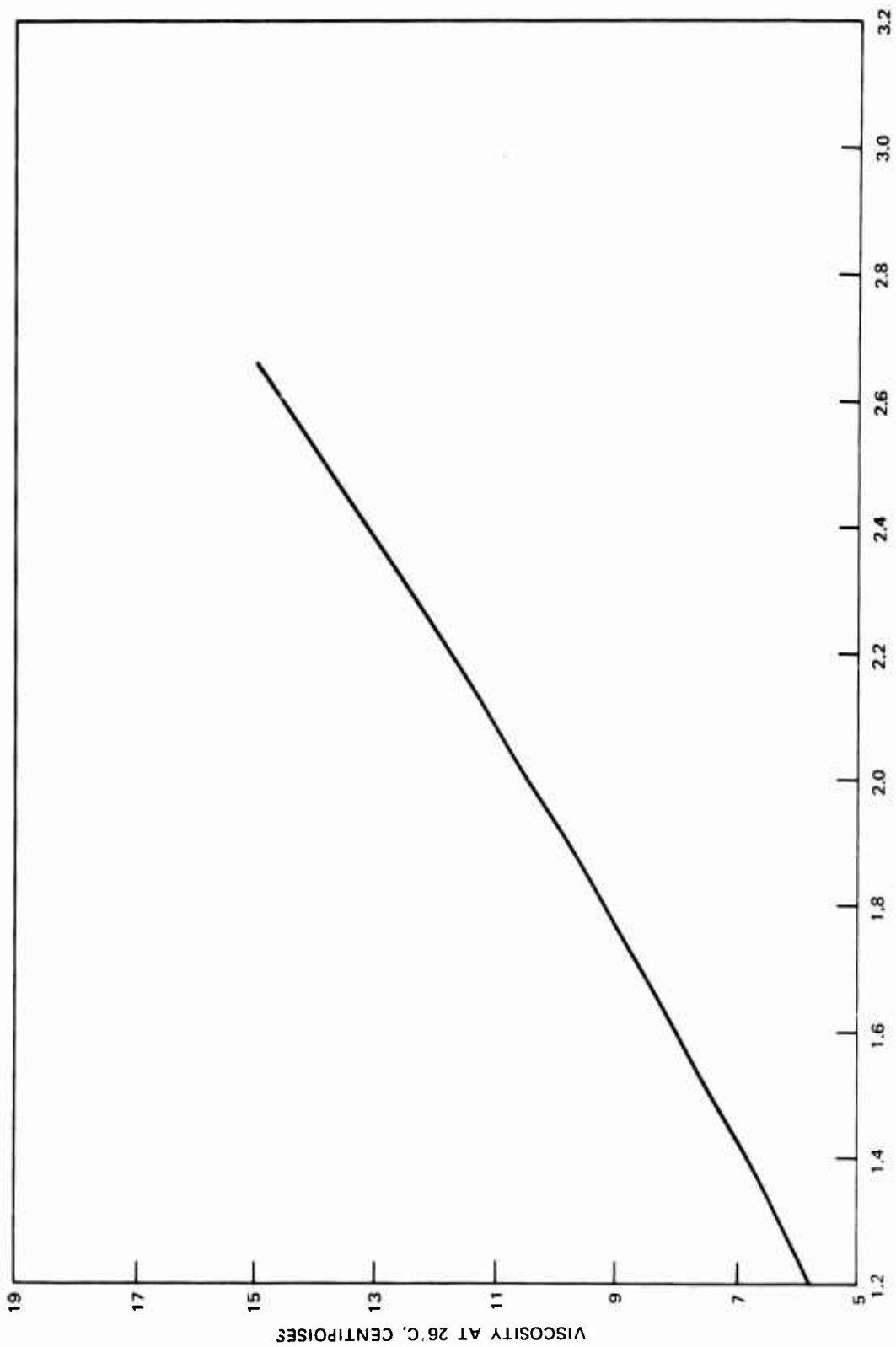


FIG. 3. Viscosity.

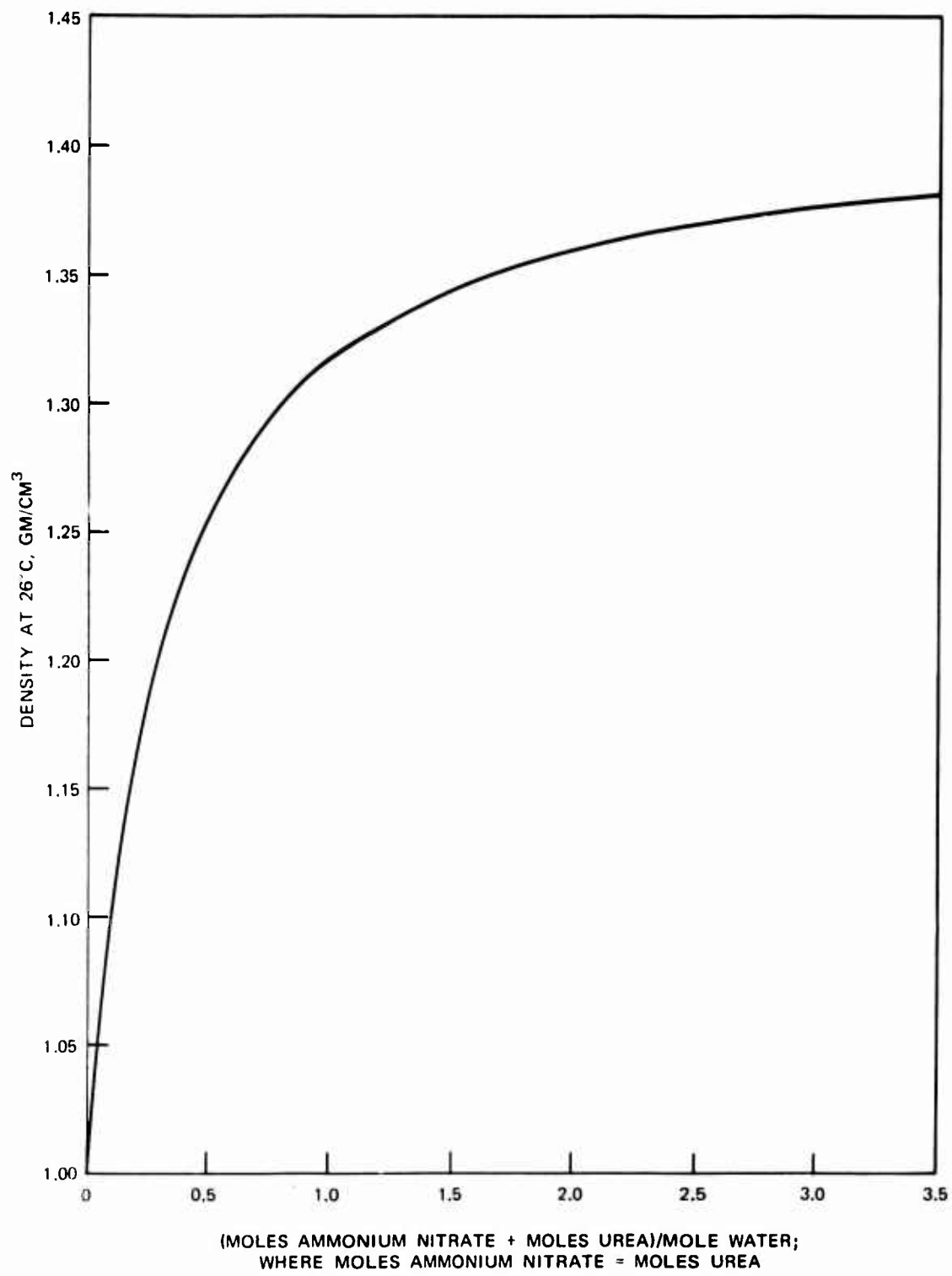


FIG. 4. Density.

TABLE 2. Corrosion of Various
Test Materials by an Equimolar
Ammonium Nitrate-Urea-Water Solution.

Test specimen	Inhibitor added to solution	Weight loss, gm/cm ² /hr × 10 ⁶
Magnesium alloy, AZ31B ^a	None	738
	1.6% thiourea and 0.8% K ₃ PO ₄	1114
Brass	None	111
	1% thiourea	8
Mild steel	None	28
	1% thiourea	0.45
Chromium plated ^b steel	None	1.3
	1% thiourea	4.7
Cadmium plated ^c steel	None	247
Aluminum	None	0
	1% thiourea	0
Lacquers and enamels	None	No visible effects

^a For tests with magnesium, the solution was diluted to 60%, the dilution in which the greatest loss occurs.
^b 0.01 mil thick.
^c 0.5 mil thick.

SENSITIVITY

Because the material contains ammonium nitrate, commonly used explosive, the NWC Propulsion Development Department was asked to conduct sensitivity data tests. The first tests were conducted with a dry mixture of ammonium nitrate with urea (4 parts nitrate, 3 parts urea). Results of these tests show that the mixture exhibits no electrostatic or friction sensitivity by the standard NWC safety tests. The samples were moderately sensitive to impact (19 cm, 2 1/2 kg weight, 50% point), much more so than ammonium nitrate tested alone (Ref. 2). However, the chemical reaction which occurred as a result of the impact of the dropped weight was not brisant. In the tests the decomposition of the dry material was non-explosive. Since the material is so hygroscopic, it will seldom, if ever, be dry. Testing with wet material (9:1 solution weight ratios) resulted in the following data:

An Olin Mathieson liquid testing apparatus was used to test the 9:1 solution for impact sensitivity. The 50% point was 158 kg-cm, which is nearly the same value as is measured when using water to calibrate the apparatus (168 kg-cm). For comparison, nitroglycerin has a value of 2 kg-cm; pentaerythritol trinitrate, a value of 8.5 kg-cm.

The detonability of the 9:1 solution was measured using the standard Naval Ordnance Laboratory (Silver Spring, Md.) card gap test. The donor charge was provided by two tetryl pellets. No cards were used to separate the 9:1 solution from the donor

(zero gap). This is the most severe test made; materials that do not detonate with zero gap will not detonate with a gap. In three out of three tests there was no detonation of the 9:1 solution. The conclusion was that 9:1 solution will not detonate. The 9:1 solution would not burn when exposed for several seconds to a burning pyrotechnic with a flame temperature in excess of 1650°C.

DROP GROWTH

The equation developed by Fukuta (Ref. 3), in which the value of the condensation coefficient, β , is 0.415 and the atmospheric pressure, P , is 1 atmosphere, was used to calculate the growth rate of droplets in moist air. Figures 5 through 10 are arranged in increasing saturation ratio, S . The maximum volume of the hygroscopic droplet, V_{\max} , is given in Fig. 5 through 8 where droplets reached maximum. Figures 5 through 9 were calculated at 20°C, a temperature likely to be encountered in warm fog; Fig. 10 was calculated at 0°C, a temperature most likely to be encountered in cold clouds. To simulate polluted fog, a "dirty" water calculation was included (Fig. 10). A polluted droplet would grow to the same size as a treatment droplet at a saturation ratio of 1.01. Figures 11 and 12 show the effect of S or relative humidity, RH, on a single initial size drop. In all of the figures, unless otherwise noted, the drop has a ratio of 2.5 moles of ammonium nitrate-urea and 1 mole of water (9:1 solution) at time zero and radius zero, r_0 . Comparisons with "dirty" water and with pure water are shown in Fig. 10 and 12.

If we assume that:

1. The calculations result in realistic values of droplet growth.
2. When droplets are less than 20 μ in radius, the probability of coalescence is small, and the fall rate is low. Therefore, when a droplet does not grow by diffusional processes to a radius greater than 20 μ , the droplet does not fall out of the fog or cloud.

We can then conclude that:

1. When $S < 0.95$ and $r_0 < 9 \mu$, then the maximum radius, $r_{\max} < 20 \mu$ and the drops are too small.
2. When $S < 0.97$ and $r_0 < 8 \mu$, then $r_{\max} < 20 \mu$ and the drops are too small.
3. When $S < 0.98$ and $r_0 < 7 \mu$, then $r_{\max} < 20 \mu$ and the drops are too small.
4. When $S < 0.99$ and $r_0 < 5.5 \mu$, then $r_{\max} < 20 \mu$ and the drops are too small.
5. When $S = 1.00$ and $r_0 = 2 \mu$, the droplets will not grow to 20 μ in 30 minutes and are too small.
6. When $S = 0.95$ and $r_0 = 25 \mu$, the droplet will gain over 1/2 of all of the water vapor it is going to gain by *diffusion* in less than 1 minute. It would take about 19 minutes to gain the other half if coalescence were not a contributing growth factor.
7. Solution droplets larger than 100- μ radius have slow growth rates and high fall velocities. They will fall to the ground in the 30 minutes or more required for them to collect water by diffusion processes alone.

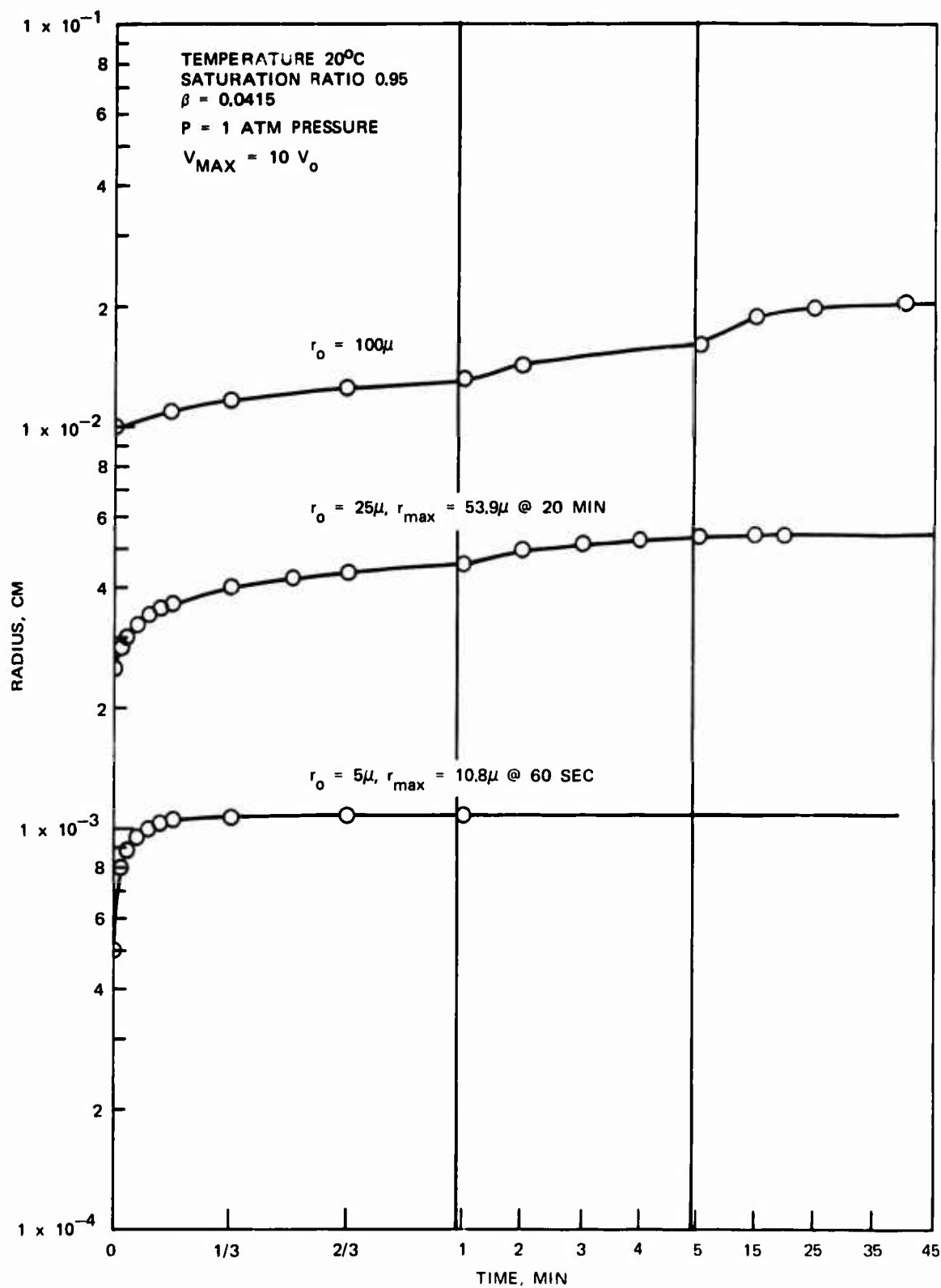


FIG. 5. Drop Growth, $S = 0.95$.

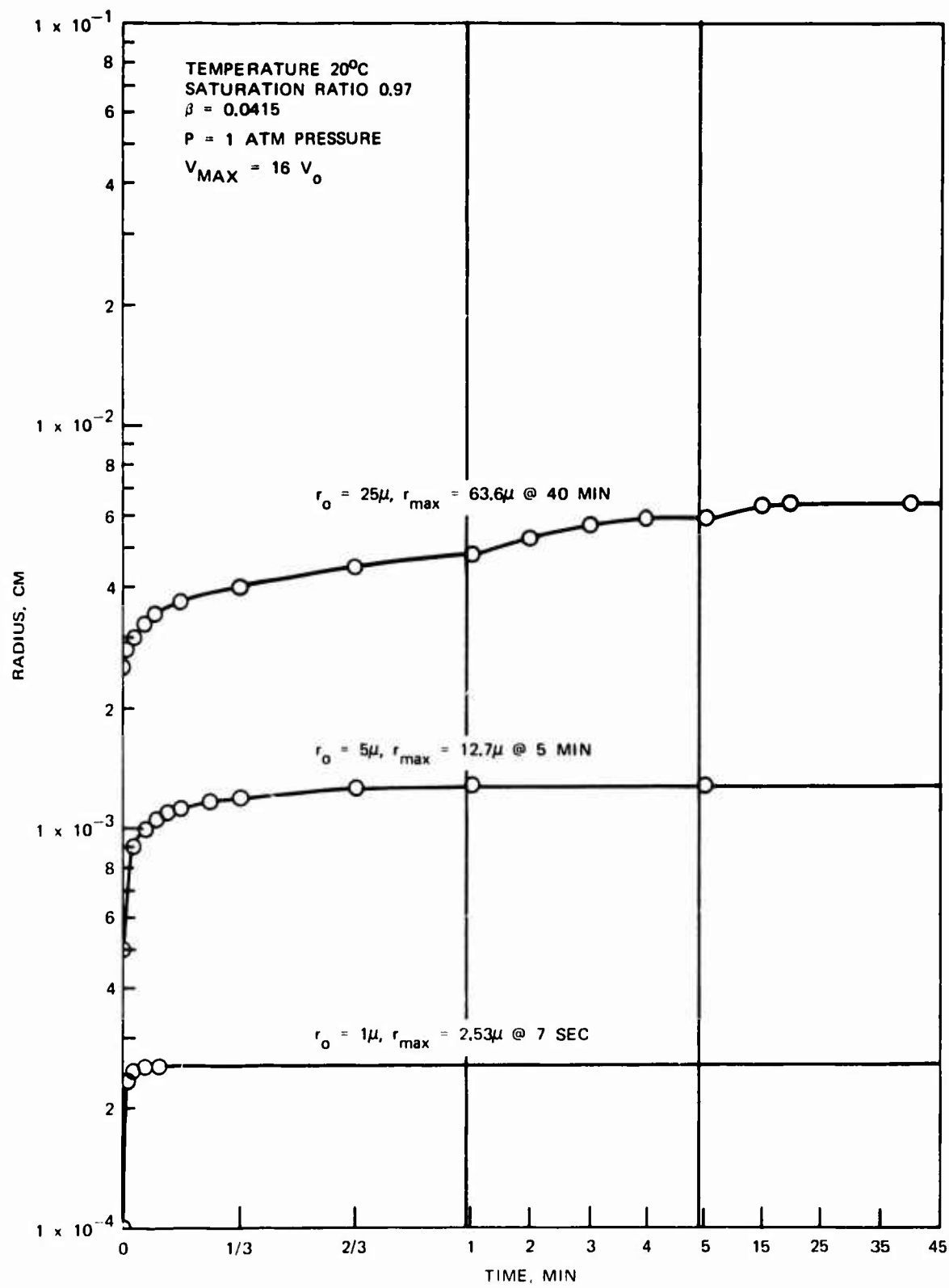


FIG. 6. Drop Growth, $S = 0.97$.

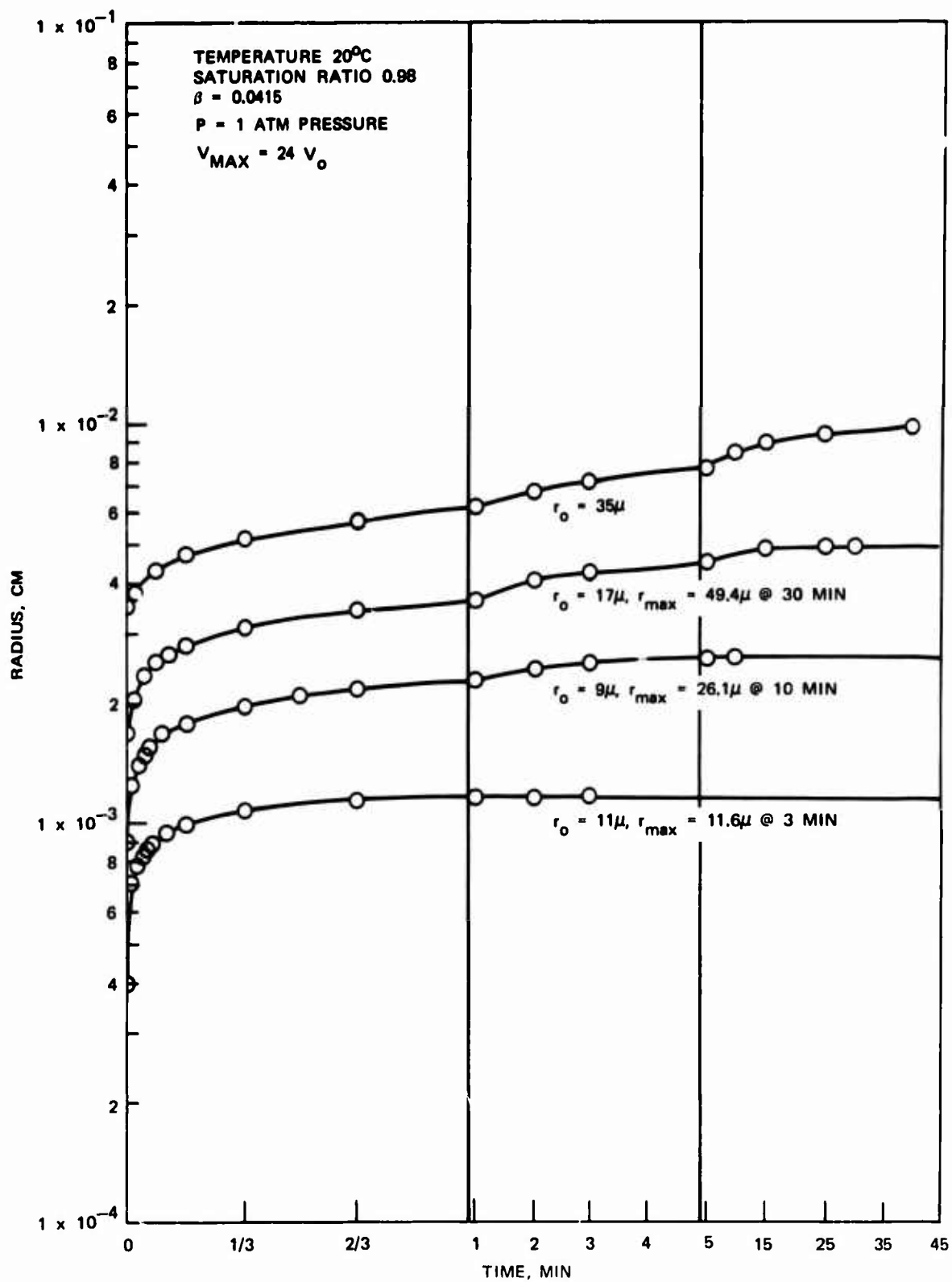


FIG. 7. Drop Growth, $S = 0.98$.

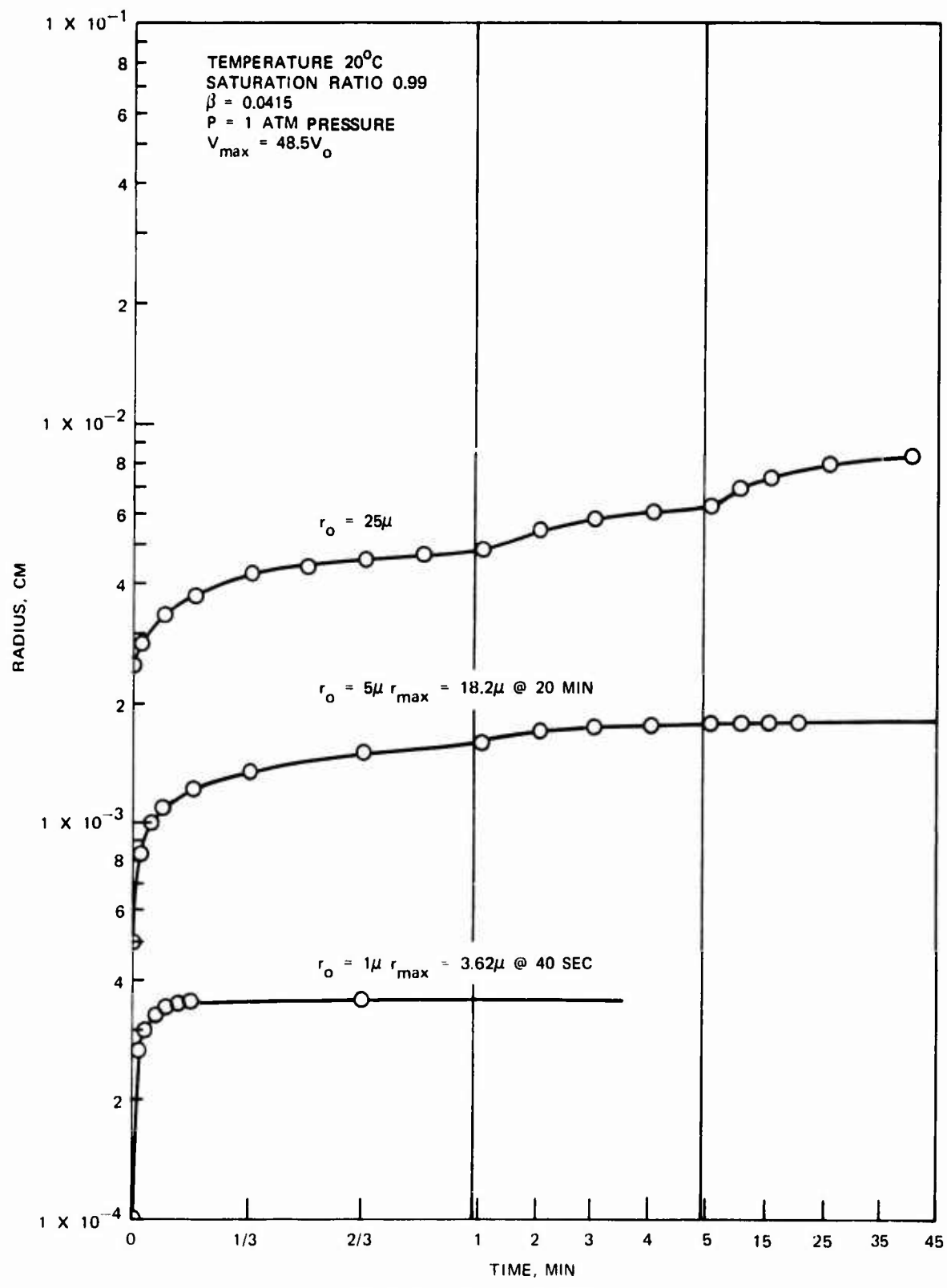


FIG. 8. Drop Growth, $S = 0.99$.

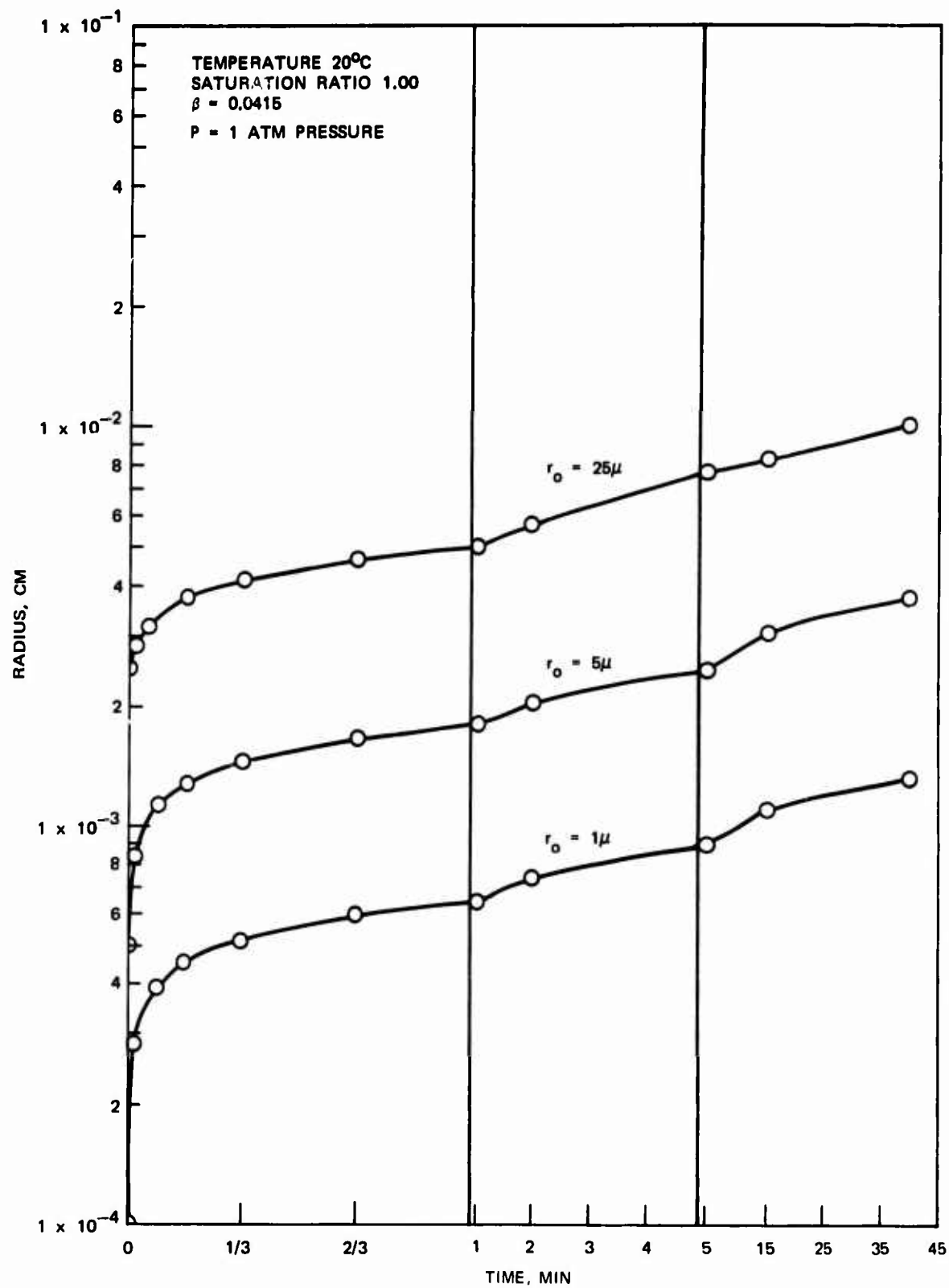


FIG. 9. Drop Growth, $S = 1.00$.

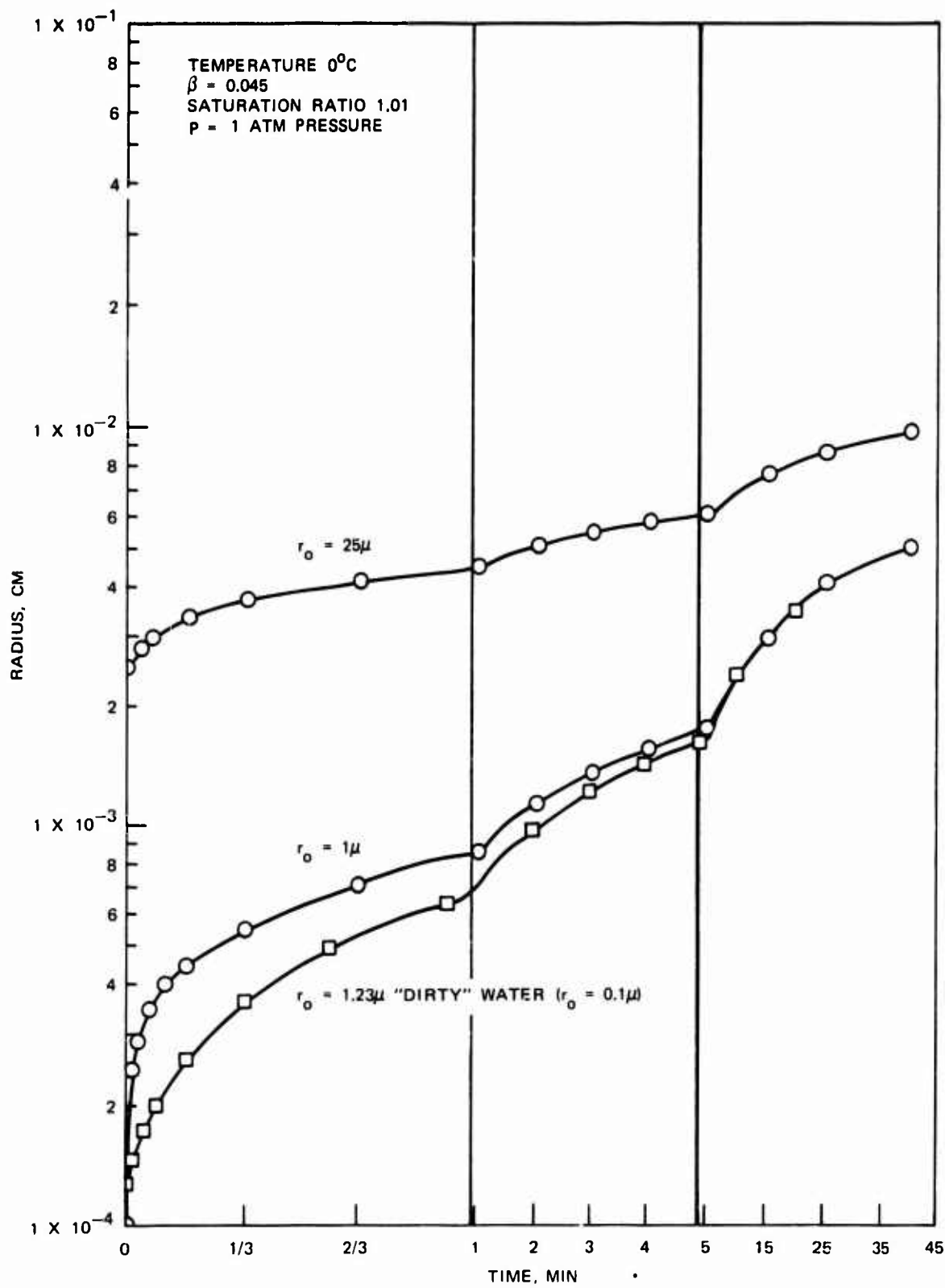


FIG. 10. Drop Growth, $S = 1.01$.

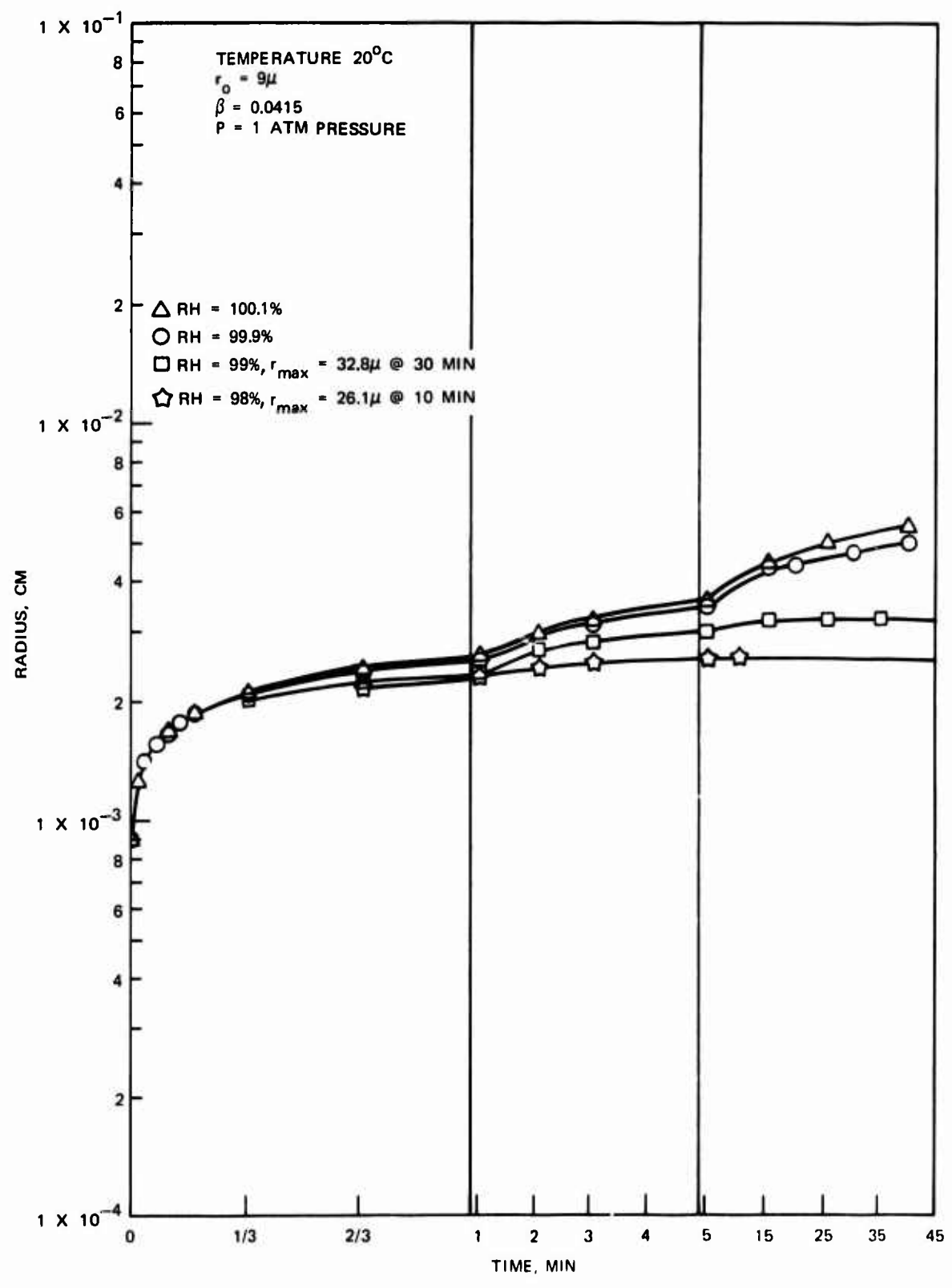


FIG. 11. Drop Growth, $r_0 = 9\mu$.

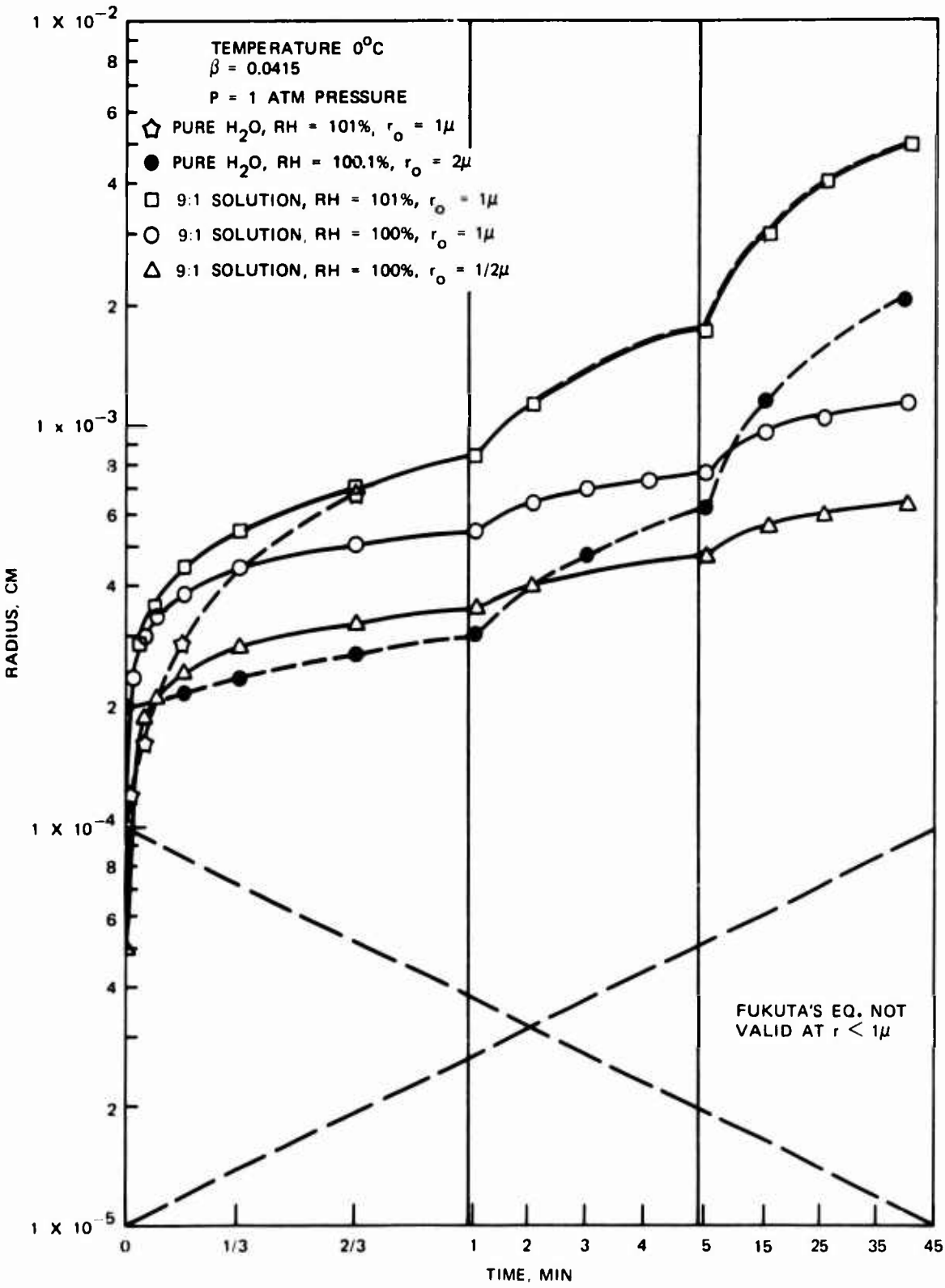


FIG. 12. Drop Growth, Agent Versus Pure Water.

8. At supersaturation and small initial droplet radii, the indigenous cloud droplets of pure water will grow to $20\ \mu$ as fast as equally small hygroscopic droplets. The upper limit of r and the lower limit of S were not calculated. At $r_0 = 1\ \mu$ and $S = 1.01$, conclusion 8 is true (Fig. 12).

9. The droplet size is important; they must not be too large or too small. The optimum size range is dependent on the atmospheric conditions.

These conclusions are in general agreement with the results of Tag and others (Ref. 4). These calculations considered the whole fog; there was a size distribution of fog and spray droplets, a varying saturation ratio, drop growth by condensation and coalescence, and vertical transfer and mixing. Tag and others (Ref. 4) concluded that the drops $35\ \mu$ and larger were too large and those $4\ \mu$ and smaller too small for fog clearing. In computer calculations in Ref. 3, an equation developed by Mordy (Ref. 5) was used to calculate the condensation growth process. When Fukuta's and Mordy's equations were compared with a singular initial drop size and at a constant saturation ratio of 0.98, the droplets grew faster with Fukuta's equation.

CONCLUSION

This highly hygroscopic agent has created no observed ecological problems. The corrosion problems have been greatly reduced and efforts to further reduce them are continuing. Finally, the cost and ease of handling, coupled with the above properties, dictate continuing studies with this material. Indeed, the saturation vapor pressure is so low and the water vapor pickup so rapid that there is little need to look further for a hygroscopic agent. While it is true that the ultimate vapor pickup by agents of lesser molecular weight and by agents which completely dissociate will be greater, the total pickup time is so long compared to the time of fall of the droplet, that the droplets will have fallen out of the fog onto the ground before they pick up the additional water. Many of the ammonium nitrate-urea droplets fall on the ground before they pick up all of the water they are capable of adsorbing.

Appendix
DROP SIZE CALCULATIONS
(Equation, Values, Computer Program, and Sensitivity of Equation)

The equation (Ref. 3) used to calculate the droplet growth was

$$\frac{dm}{dt} = 4\pi r_s' (S - A) \left[\frac{AMLL'}{RT_\infty^2 K f_{3\alpha}} + \frac{RT_\infty}{D M p_{\infty, sat} f_{3\beta}} \right],$$

where

$$A = \frac{n}{n + in'} \exp \left[\frac{2\sigma' V_L'}{r_s' RT_\infty} \right]$$

where

- m = mass of droplet
- t = time
- r_s' = radius of solution droplet
- S = saturation ratio
- A = combination of Kelvin and solute effects, defined by Fukuta
- M = molecular weight of water
- L = latent heat of condensation of water vapor
- L' = latent heat of condensation of water vapor in solution
- R = universal gas constant
- T_∞ = ambient temperature
- K = thermal conductivity of air
- $f_{3\alpha}$ = correction factor for temperature difference, defined by Fukuta
- D = diffusion coefficient of water vapor in air
- $p_{\infty, sat}$ = saturated water vapor pressure at ambient temperature, T_∞
- $f_{3\beta}$ = correction factor for vapor density difference, defined by Fukuta
- n = number of moles of solvent, that is, water
- n' = number of moles of solute
- i = van't Hoff factor for non-ideal solution
- σ' = surface tension of solution droplet
- V_L' = molar volume of water in solution

In the ammonium nitrate-urea-water solution, the ammonium nitrate and urea are in equal mole ratios. Initially, the ratio of moles of ammonium nitrate and urea is 2.5.

Fukuta's equation for the growth of a hygroscopic drop was programmed on a computer using EXFOR language on the International Time Sharing Corporation computer. The units of measure, grams, centimeters, and seconds, were used to dimension the variables and constants in the equation. The correction factors $f_{3\alpha}$ and $f_{3\beta}$ were calculated using equations taken from Fukuta's report (Ref. 3).

$$f_{3\alpha} = \frac{r_s'}{r_s' + \frac{0.22 \times 10^{-4}}{P} \sqrt{\frac{273}{T_\infty}}}$$

where

P = atmospheric pressure

$$f_{3\beta_1} = \frac{r_s'}{r_s' + 3.9 \times 10^{-4} \left(\frac{D}{0.22}\right) \sqrt{\frac{273}{T_\infty}}}$$

$$f_{3\beta_2} = \frac{r_s'}{r_s' + 8.1 \times 10^{-4} \left(\frac{D}{0.22}\right) \sqrt{\frac{273}{T_\infty}}}$$

$f_{3\beta_1}$ differs from $f_{3\beta_2}$ because the condensation coefficient, β , is varied from 0.0415 to 0.02.

Some of the independent variables were in the program in tabular form. Table 3 gives van't Hoff factors, i , density, ρ_L' , and surface tension, σ' , as a function of concentration of solute in a drop, n'/n . Table 4 gives latent heat of condensation, L , latent heat of condensation of water vapor in solution, L' , and saturated water vapor pressure, $P_{\infty sat}$, as a function of ambient temperature, T_∞ .

TABLE 3. Van't Hoff Factors, Density, and Surface Tension That Vary as a Function of Concentration of Solute in Drop.

n'/n	i at 26°C	ρ_L' at 26°C, gm/cc ^a	σ' at 20°C, gm/cm ^b
2.5	1.07	1.370	84.8×10^{-3}
2.0	1.10	1.360	84.5
1.5	1.18	1.345	84.1
1.0	1.38	1.316	83.3
0.5	1.65	1.253	81.5
0.4	1.70	1.230	80.9
0.3	1.70	1.200	79.9
0.2	1.63	1.160	78.5
0.1	1.65	1.100	76.4
0.05	1.50	1.050	74.9

^a ρ_L' = density of solution droplet.
^b σ' = surface tension of solution droplet.

TABLE 4. Latent Heat and Vapor Pressure That Vary as a Function of Temperature.

T_∞ , °C	L , gm cal/gm	L' , gm cm/gm	$P_{\infty sat}$, gm/cm ²
0	596	$2.53 \cdot 10^7$	6.2
10	590	$2.51 \cdot 10^7$	12.5
20	585	$2.49 \cdot 10^7$	24.8

The other constants and variables are listed below along with their units of measure.

$$D = 0.22 \left(\frac{T_{\infty}}{273} \right)^{1.74} / P, \text{ cm}^2/\text{sec}$$

$$K = 5.57 \times 10^{-5}, \frac{\text{gm cal cm}}{\text{sec cm}^2 \text{ } ^\circ\text{C}}$$

m = mass, grams

m_0 = initial mass of droplet (at $n'/n = 2.5$), grams

M = 18 gm/gm mole

$$n = \frac{[4/3\pi(r_s')^3 \rho_L'] - 70n'}{18}, \text{ moles}$$

$$n' = \frac{m_0(175/193)}{70}, \text{ moles}$$

P = pressure, atm

$$r_s' = \sqrt[3]{\frac{m}{4/3\pi\rho_L'}}, \text{ cm}$$

r_{so}' = initial radius, cm

$$R = 8.314 \times 10^4, \frac{\text{gm cm}}{\text{gm mole } ^\circ\text{K}}$$

S = saturation ratio, dimensionless

t = seconds

T_{∞} = 273, 283, or 293°K

$$V_L' = 17.755, \frac{\text{cc}}{\text{gm mole}}$$

Note: An average value of V_L' was used, since it varied little and had a small effect on calculations.

The computer program is given in Table 5.

TABLE 5. Computer Program for EXFOR Language.

```

100 REAL I,M,K,N,MDOT,MDOTP,I,L,LP,NP,NON,MDOTQ
110 DIMENSION NON(12),I(12),RHO(12),SIG(12),SS(12)
120 DIMENSION L(3),LP(3),PIS(3),T(3)
130 OPEN (2) INPUT,JOHN
140 T(1)=273.; T(2)=283.; T(3)=293.
150 L(1)=-.596.; L(2)=-.590.; L(3)=-.585.
160 LP(1)=-2.53E7; LP(2)=-2.51E7; LP(3)=-2.49E7
170 PIS(1)=6.2; PIS(2)=12.5; PIS(3)=23.8
180 CR=8.314E4
190 M=18.
195 DT=.02; TIME=0.
200 K=5.57E-5
205 MDOTQ=0.
210 D=.22*(T(I)/273.)↑1.74/P
220 PI =3.14159

```

TABLE 5. (Contd.)

```

230 DO 9 JJ=1,10; J=11-JJ
240 9 READ (2,0) NON(J), I(J),RHO(J),SIG(J)
250 1 WRITE (1,0)"INPUT IT, IB,S,R,P"
260 READ (0,0) IT,IB,S,R,P
270 YM=1.333333*PI*R13*1.37
280 RHOI=1.37
290 DO 99 LIM=1,500
291 IF (TIME > .199) DT=.1
292 IF (TIME > .99) DT=1.
293 IF (TIME > 9.99) DT=5.
294 IF (TIME > 29.99) DT=10.
295 IF (TIME > 59.99) DT=60.
297 IF (TIME > 599.9) DT=300.
298 IF (TIME > 1799.9) DT=600.
300 YMZ=YM
310 DO 79 J=1,25
315 RS =(YM/1.33333*PI*RHOI))†(.33333333)
320 NP=1.333333*PI*R13 *(1.37/70.)*(175./193.)
340 N =(1.333333*PI*RS13*RHOI - 70.*NP)/18.
350 X =NP/N
360 II=QUIN(X,I,NON,1,10,2,SS)
365 IF (X < .05) II=AMAX(II,1.5)
370 RHOI=QUIN(X,RHO,NON,1,10,2,SS)
372 IF (RHOI < 1.) RHOI=1.
375 VM=17.755
380 SIGI=QUIN(X,SIG,NON,1,10,2,SS)
385 IF (SIGI < 74.2E-3) SIGI=72.4E-3
390 F3A=RS/(RS+ 000022/P*SQRT(T(IT)/273.))
400 IF (IB .EQ. 1) F3B=RS/(RS +.00039*D/.22 *SQRT(273./T(IT)))
410 IF (IB .EQ. 2) F3B=RS/(RS +.00081*D/.22 *SQRT(273./T(IT)))
420 A=N/(N+II*NP) *EXP(2.*SIGI*VM/(RS*CR*T(IT)))
430 VA=A*M*LP(IT)*L(IT)/(CR*T(IT)†2*K*F3A)
440 VB=CR*T(IT)/(D*M*PIS(IT)*F3B)
450 MDOT=4.*PI*RS*(S-A)/(VA+VB)*.5 +.5*MDOTQ; IF (J .EQ. 1) GO TO 78
451 IF (ABS(MDOT-MDOTQ)/(ABS(MDOT)) < .000001) GO TO 80
452 78 MDOTQ=MDOT
460 IF (LIM .NE. 1 .OR. J .NE. 1) GO TO 79
462 MDOTP=MDOT
463 LIP=0
464 WRITE (1,2)LIP,YMZ,RS
470 79 YM=YMZ+.5*(MDOT+MDOTP)*DT
475 WRITE (1,0)"HARK, NO CONVERGENCE!",MDOT,MDOTQ
478 80 YM=YMZ+.5*(MDOT+MDOTP)*DT
480 MDOTP=MDOT
483 TIME=TIME+DT
487 LIP=TIME+.01
489 IF (LIP < 1) GO TO 99
490 WRITE (1,2)LIP,YM,RS
491 99 CONTINUE
495 2 FORMAT (/ "T=",14," M=",E12.4," RS=",E12.4)
500 GO TO 1
510 END
1000 FUNCTION QUIN(X,G,XL,N,M,L,S)
1010C LINEAR (L=1) AND QUADRATIC (L=2) INTERPOLATION ROUTINE

```

TABLE 5. (Contd.)

```

1020C X IS THE INPUT POINT; G(N,I) VS. XL(I) IS THE CURVE TO BE
1030C INTERPOLATED. M IS THE NUMBER OF POINTS IN XL(I).
1040C S(I),I=1,M ARE THE INTERPOLATION COEFFICIENTS.
1050C NOTE THAT IF N IS LESS THAN OR EQUAL TO ZERO ONLY THE IN-
1060C TERPOLATION COEFFICIENTS (S) ARE COMPUTED AND G IS NOT USED.
1070 DIMENSION G(1,12),XL(12),S(12)
1080 NMM=M-1; K=NMM
1090 DO 1 I=1,M
1100 1 S(I)=0.
1110C THE X-LATTICE (XL) VALUES MUST BE IN ASCENDING ORDER.
1120 DO 2 J=1,NMM
1130 IF (X .GT. XL(J+1)) GO TO 2
1140 K=J
1150 GO TO 3
1160 2 CONTINUE
1170 3 S(K+1)=(X-XL(K))/(XL(K+1)-XL(K))
1180 S(K)=(XL(K+1)-X)/(XL(K+1)-XL(K))
1190 IF (L .EQ. 1) GO TO 6
1200 W1=0.
1210 IF (K .EQ. NMM) GO TO 4
1220 YF0=-1./(XL(K+1)-XL(K))
1230 YF2=-1./(XL(K+2)-XL(K+1))
1240 YF1=-YF0-YF2
1250 W1=X-XL(K)
1260 W2=0
1270 4 IF (K .EQ. 1) GO TO 5
1280 YB0=-1./(XL(K)-XL(K+1))
1290 YB2=-1./(XL(K-1)-XL(K))
1300 YB1=-YB0-YB2
1310 W2=XL(K+1)-X
1320 5 VA=W1+W2
1330 W1=W1/VA*(X-XL(K))*(X-XL(K+1))/(XL(K)-XL(K+2))
1340 W2=W2/VA*(X-XL(K+1))*(X-XL(K))/(XL(K+1)-XL(K-1))
1350 IF (K .GT. 1) S(K-1)=W2*YB2
1360 S(K)=S(K) +W1*YF0 +W2*YB1
1370 S(K+1)=S(K+1) +W1*YF1 +W2*YB0
1380 IF (K .LT. NMM) S(K+2)=W1*YF2
1390 6 QUIN=0.
1400 IF (N .LE. 0) GO TO 99
1410 DO 7 I=1,M
1420 7 QUIN=QUIN+S(I)*G(N,I)
1430 99 RETURN
1440 END
READY

```

The calculated sizes are very precise and are not in error by more than a few percent. The calculated times can be in error by one or two integration steps. The integration steps used were

<i>Time, sec</i>	<i>Integral time, sec</i>
$t < 0.2$	$\Delta = 0.02$
$t > 0.2$	$\Delta = 0.1$
$t > 1.0$	$\Delta = 1$
$t > 10$	$\Delta = 5$
$t > 30$	$\Delta = 10$
$t > 60$	$\Delta = 60$
$t > 600$	$\Delta = 300$
$t > 1,800$	$\Delta = 600$

Calculations other than those shown in Fig. 5 through 12 indicate the following to be true of Fukuta's equations when used on ammonium nitrate-urea-water:

1. Use of a condensation coefficient, β , of 0.02 in place of 0.0415, causes at most, a 5% change in calculated radius. The maximum change in radius occurs when $r_0 = 1 \mu$ (r_0 is the radius of the initial ammonium nitrate-urea droplet). With $S = 1$, the difference in radius decreases with time.

2. Using 20°C as a base temperature, the use of 10°C causes an 8% maximum decrease in calculated radius; and the use of 0°C causes a 17% maximum decrease. The maximum change in radius occurs when $r_0 = 1 \mu$. With $S = 1$, the difference decreases with time.

3. Since the diffusion coefficient increases with altitude, growth rate increases too. Using sea level as a base, at 4 km a 1- μ droplet grows 10% more at 300 seconds elapsed time; at 6 km, 15% more. The maximum difference occurs when $r_0 = 1 \mu$. With $S = 1.01$, the difference increases with time.

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